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PLUTONIUM DISTRIBUTION IN ROCKY FLATS SOIL*

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Abstract—Plutonium concentrations in Rocky Flats soil were inversely proportional to distance from the plutonium source, to depth of the sample, and to particle size of sieved soil samples Coefficients of variation ranged to more than 300%, and frequency distributions of plutonium concentrations in samples were highly skewed The plutonium distribution patterns and known characteristics of the plutonium source indicated that the mechanisms of environmental dispersion may have involved the attachment of plutonium oxide to soil particles; primary dissemination of contaminant from the source by wind, and weathering, microdispersal, and penetration into soil of deposited particles The high degree of spatial variability, in particular, suggested that the most common functional form of the contaminated soil during dissemination was probably an agglomerated particle containing many plutonium oxide and soil particles bound together

INTRODUCTION

A COSTLY fire at Rocky Flats in 1969 and later detection of off-site ^{239}Pu in soil samples (Ma70; Po72) spurred discussion of the environmental contamination problems and safety implications of the Rocky Flats installation (Jo76; Kr70; Ma70; Po72; Sh71) Further investigations indicated that the primary contaminating event was leakage from drums containing plutonium-laden cutting oil that had been stored outdoors in the southeast corner of the plant (Kr70; Po72).

Soil is the most important ecosystem compartment at Rocky Flats with regard to fraction of total plutonium contained and potential for plutonium transport (Li76) Consequently, this paper examines data from a study, begun in late 1971, of the patterns of plutonium contamination in Rocky Flats soil Specifically, this report describes (1) data on plutonium concentrations in soil, (2) the relationship of concentration to location, depth, and soil particle size, and (3) a description of the likely contamination mechanisms

The Rocky Flats (RF) installation, operated by Rockwell International for the Energy Research and Development Administration (ERDA), is located about 12 km, northwest (NW) of the nearest portions of the Denver, Colorado metropolitan area at an elevation of over 1800 m ERDA controls approximately 30 km^2 of land, most of which is used as a buffer zone to separate the public from plant production operations.

Topography of the installation is characterized by a series of flat, wind-scoured plateaux divided by five separate watercourses running roughly from west to east Rocky Flats climate is typified by strong and often gusty winds (3 7 m/sec mean) and moderate rainfall (40 cm/yr mean). Typically, the stronger winds at RF blow from the west (W) and NW, during 1975, 22% of the recorded winds were from the NW (An76). Vegetation at the installation is modified grassland. Ex-

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cept for a new buffer zone purchased in 1975, Rocky Flats land has not been significantly disturbed by man for over 25 yr. The flora and fauna have been described previously (We74; Wh73).

Data from RF air sampling station S-8 (Hu76), located about 75 m southeast of the barrel storage area, agree with information gathered by ERDA's Health and Safety Laboratory (Kr70) that identifies the barrel storage area as the source of the east-southeast contamination pattern at the site. Monthly averages of daily air samples indicated that gross alpha activity was associated with periods of known perturbation of the contaminated surface (Table 1, Fig. 1). These data indicate the time of the original plutonium dispersal and provide strong evidence that the barrel storage area was the main source of Pu contamination in the downwind ecosystem until the area was covered by asphalt in 1969.

*Table 1 Total monthly gross alpha activity in ambient air at station S-8 (75 m east of oil barrel storage area) during perturbation events of the storage area surface**

Dates	Event	Alpha activity (dis/min/m³)
7/59–9/63	No large scale leaking	0 020
1/64–1/65	Large scale leaking	0 056
1/65	Contaminated soil covered with fill	0.022
1/66	Small building added to filter contaminated oil from leaking to new drums	0 031
1/67	Drum removal activity begun	0 084
6/68	Last drums removed but high winds spread some activity	0.417
2/69	Weeds burned area graded for paving	0.067
9/69	Asphalt pad completed	0.029
11/69	Four sampling wells dug through pad	0.073
4/71	Drainage ditch dug on west side of asphalt pad	0 073

*Air filter material was counted directly in a gas-flow proportional detector Data adapted from Hu76.

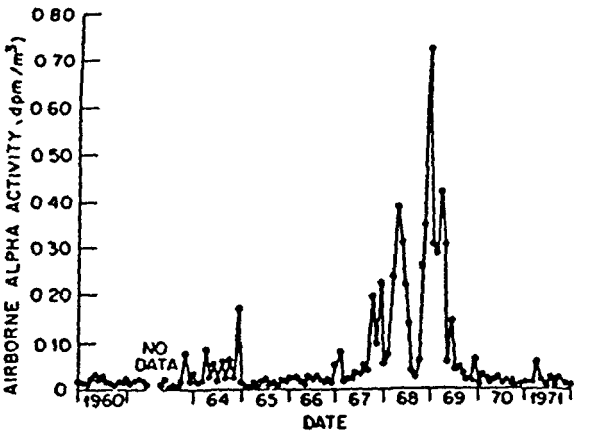

FIG 1 Monthly means of daily gross alpha activity in ambient air at station S-8 (75 m east of the oil barrel storage area) Aliquots of Gelman AE filter material were counted in a gas-flow proportional detector Data adapted from Hu76

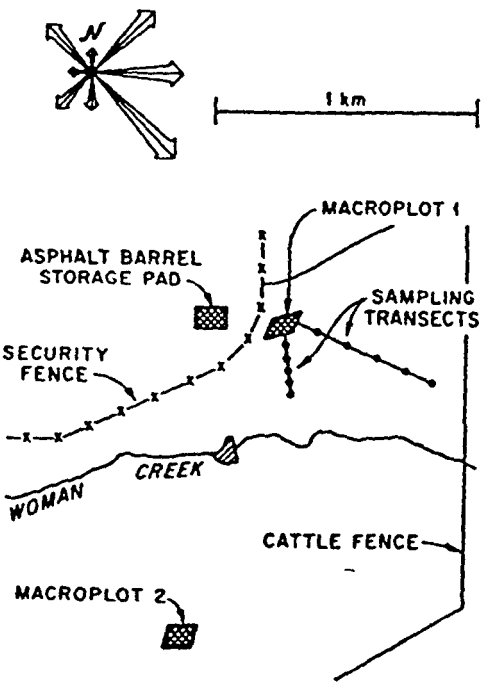

FIG 2 Schematic representative of southeast corner of Rocky Flats installation showing location of Macroplot 1 and two sampling transects in relation to barrel storage area The wind rose indicates the directions toward which the mean winds blew during 1974

METHODS AND MATERIALS

We established two macroplots for study (Fig. 2). The Macroplot 1 sampling grid covered about 0.58 ha and contained 100 microplots (grid intersections). The Macroplot 2 grid had 25 microplots and covered about 0.12 ha Macroplot 1 presumably had the highest concentrations of Pu in soil outside the security fence and had a reasonably undisturbed vegetative community Conversely, Macroplot 2, by virtue of distance, direction, and the presence of sheltering topography between the source and the macroplot, was presumed to be nearly background Vegetative communities of the two

macroplots were similar. During 1972–1974, four replicate soil samples from each 3-cm increment from 0 to 21 cm were taken at ten and five randomly chosen sample microplots (intersections) on Macroplot 1 and Macroplot 2, respectively.

We established two sampling transects: one extending 500 m east-southeast (ESE) from the eastern boundary of Macroplot 1 and the other running 250 m approximately south (S) from the southern boundary of Macroplot 1 (Fig 2). Four soil samples 0–3 cm were taken at each 100-m and 50-m interval along the ESE and S transects, respectively. Using aerial photographs, we estimated the distance east (X) to each sample location from a north–south line through the center of the asphalt pad and, similarly, the distance south (Y) from an east–west line to each sample of Macroplot 1 and the two transects.

Soil samples were air dried, and material greater than 0.5 cm in diameter was removed from the sample. After drying the samples in an oven, they were weighed and mechanically shaken on brass soil sieves with meshes ranging from 2000 to 45 μm, as listed in Table 2. The accumulation on each sieve and the underlying pan was weighed and sealed into a paper envelope.

Soil samples weighing approximately 5 g were sent to commercial laboratories (LFE Environmental, Richmond, California, and Eberline Instrument Corp., Albuquerque, New Mexico) or analysed in our laboratory LFE Environmental used concentrated hydrofluoric acid (We71) and Eberline modified a pyrosulfate fusion technique to dissolve samples (Si69) Ion exchange columns were used to remove interfering nuclides and to isolate plutonium from the samples before counting by alpha spectrometry. Chemical recovery was measured by addition of ^{236}Pu tracer to each sample (We71; Si69) Agreement between homogenized split samples sent to these laboratories was good (Li76). In our laboratory, a procedure incorporating harsh digestion with HNO_3 and HF, ion exchange, organic extraction, and liquid scintillation (LS) counting was used and had an estimated minimum detectable concentration of 0 18 dis/min/g for 5-g samples (Li76) Plutonium data are ^{239,240}Pu unless otherwise noted

RESULTS

Surface soil samples had a higher mean concentration than the samples below the surface (Table 2), and Macroplot 1 had a higher mean Pu concentration than Macroplot 2 Variations in soil Pu concentrations, with regard to depth, particle size distribution, and spatial dispersion, were large in samples from both macroplots (as great as 2 0 in Macroplot 1 and 4 0 in Macroplot 2) In one case, three adjacent soil columns (5 × 5 cm) from a 5 × 15 cm area of Macroplot 2 had mean plutonium concentrations of 1060 (column A), 11 9 (column B), and 1 26

Table 2. Mean plutonium concentrations and coefficients of variation (CV = standard deviation ÷ mean) of soil samples from two Rocky Flats macroplots. The number of samples totaled 931

Macroplot	Soil particle size range (μm)	Plutonium concentration dis/min/g [mean CV] of depth group aliquots 0–3 cm	3–6 cm	6–9 cm	9–12 cm	12–15 cm	15–18 cm	18–21 cm	0–21 cm
1	2000–850	1600, 2 0	300, 1 4	200 1.8	59 0.8	30 1.6	12, 0 9	4 0 0.6	3^0 3.9
	850–425	1000 0.8	270, 0 8	230 1 7	81 1.0	65 0 7	16, 0.9	12 0.5	290 1.8
	425–250	980 0.6	290, 0.8	200 1.8	66 0.5	66 0.5	29 0 8	13 0 2	300 1.6
	250–180	1000 0.5	280, 0.6	290 1 4	86 0 9	66 0.5	31 1 1	20 0 7	310 1.5
	180–150	1700 1 1	280, 0.6	230 1 7	77 0.6	310 1.6	57 0.7	14 1 1	450, 2.1
	150–75	1900, 0 7	460 0.8	230 1 4	150 0.9	120 0 7	97 1.3	24 0 9	530 1 7
	75–45	3100 0.5	700, 0.5	460 1 4	220 0.6	360 0.7	190 1 4	79, 0 6	880 1.5
	45–0	3300 1.6	400 1 1	1800 1 8	430 1.0	500 1.0	190 1.3	60 0.3	1300 2.4
	<2000	1900, 1.5	360, 0 9	590 2.8	150 1 4	190 1.6	75 1.8	27 1 1	570 2.7
2	2000–850	13 1 7	2.1, 0.9	0.8 1.6	1.2, 1.6	0 7 1.3	0 8 1.5	1.3 0.8	3.2, 3 1
	850–425	12, 1 7	1.5, 0 9	1.0 1.5	1.2, 1.6	0.8 1 4	0.9 1.6	2.7 0 6	3.2, 3.0
	425–250	30 1 9	4.5 1 1	3 0 1.2	3 4 1.6	3 2, 1.5	3 4 1.5	8.1 0 7	8.7 2.9
	250–180	59 2.3	12, 1 1	3 1 1.5	4.5 1 4	8 7 1 4	5.8 1 4	9.5 1 1	17 3.5
	180–150	110, 2.8	22, 2.0	5.8 1.3	7 0 0 9	8.2, 1 6	8.9 1 1	11 1 0	28 4 7
	150–75	150, 3.3	4.2, 1 1	3 9 1 4	3 9 1 1	4 7 1.6	6.1 1.3	7 9 0 8	31, 6.7
	75–45	340, 3.2	13, 1.0	6.7 1 4	6 0 1.2	12, 1.9	29 1 7	12, 0 9	69 6.5
	45–0	29 1.3	7 4 1 1	9 0 1 4	7 1 1.0	8 0 1 6	25 1 4	11 0.5	14, 1.6
	<2000	92, 4.6	8 1 2.1	4 5 1 6	4 2, 1.3	5.8 2.0	9.3 2.3	8 0 1 0	22, 8.4

(column C) dpm/g at the 0–3 cm depth. Differences between the three columns extended below the surface as well, but the pattern shown in the 0–3 cm depths did not hold. Virtually all of the plutonium in column A was found in the top 3 cm, the other depths being at or near background. In columns B and C, however, the majority of the plutonium was found at lower depths.

Generally, radionuclide contamination of the environment results in log-normal distributions (Wh66; Ce69; Pi75). Following that pattern, plutonium data from soil sampling were highly skewed (Sn67, $P < 0.05$). However, the natural log transformation of these data did not result in normal distributions [as judged by the Kolmogorov–Smirnov one-sample test, (Si56)], but did generally reduce the skewness for the seven depth groups from each macroplot tested

Linear, exponential, and power function regressions of Pu concentrations in the surface samples as functions of X or Y distance from the asphalt pad were calculated. The power function was significant ($P < 0.01$) and gave the best fits of the data for both curves (Figs 3 and 4) Based on a t-test (Dr66), the slope of the Y curve (Fig 4) was significantly steeper ($P < 0.05$) than the X curve (Fig. 3). Of several multiple linear regression models attempted, the one accounting for the largest amount of the total variation, 86.8% had the following parameters:

$$\ln \mathrm{Pu} = 24.76 - 0.1187 \ln X - 3.615 \ln Y,$$

where Pu = Pu concentration (dis/min/g), X = distance east of asphalt pad centerline (m) and Y = distance south of asphalt pad centerline (m).

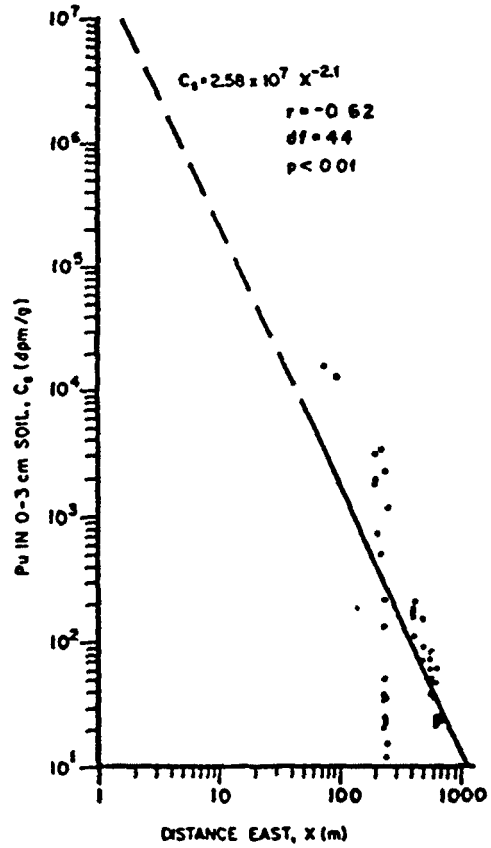

FIG 3. Plutonium concentration in 0–3-cm Rocky Flats Macroplot 1 soil as a function of distance east of the center of the asphalt oil barrel storage pad.

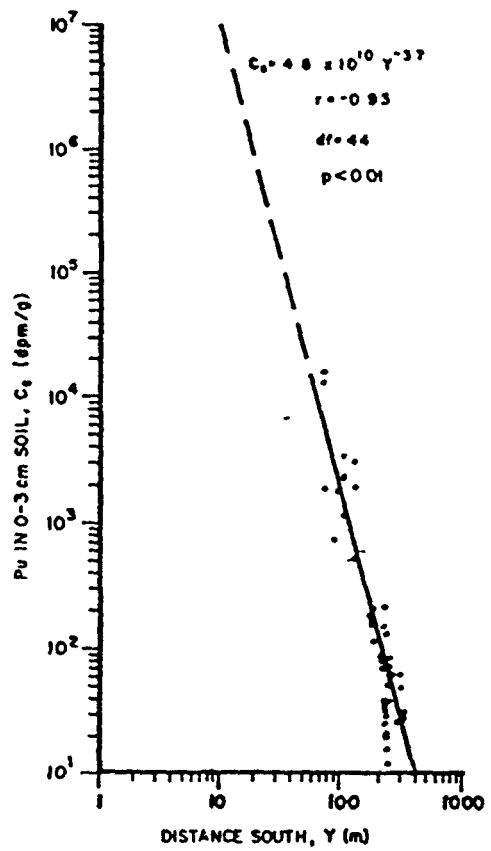

FIG 4 Plutonium concentration in 0–3-cm Rocky Flats Macroplot 1 soil as a function of distance south of the center of the asphalt oil barrel storage pad

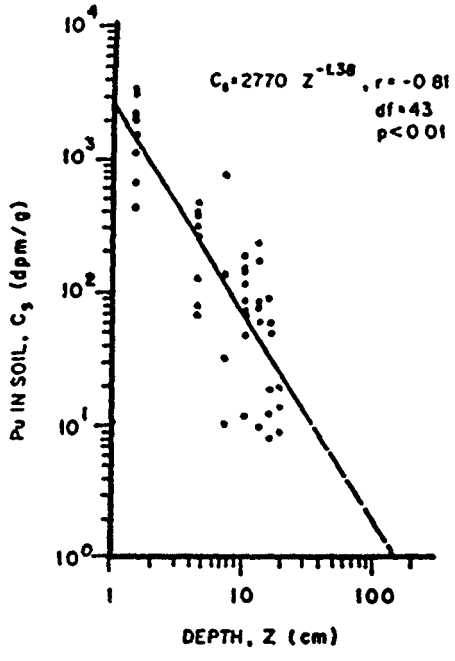

FIG 5 Plutonium concentration in Rocky Flats Macroplot 1 soil as a function of depth of sample. Sample concentrations corrected for distance east and south of center of asphalt oil barrel storage pad

Using this model, Pu concentrations of samples in the soil depth profile were adjusted to the expected concentration at a common location. The adjusted values were then regressed as a function of the sample depth (Fig. 5). The power function form of the relationship was significant ($P < 0.01$) and had the highest correlation of Pu concentration with depth of the models attempted.

The relationship of plutonium concentration in Macroplot 1 soil vs soil particle diameter (as represented by the opening of the final passage sieve) was examined for each depth using linear, exponential, and power function models. The following model most often gave significant results: ln Pu concentration = $b_0 + b_1$ ln diameter (Table 3). The steepest slope, at the 12–15-cm depth, was significantly different from the flattest slope, at the 3–6-cm level ($P < 0.05$). However, there seemed to be no clear-cut trend in slope of the Pu vs soil-particle size relationship with depth.

A tabulation of the sieve fraction data by size range and depth for both macroplots did not produce any particular pattern with either depth or particle size range Furthermore, regressions of fraction of total soil mass per sample as a function of depth were not significant for most sieve fractions.

Table 3 Regression parameters of soil plutonium concentration (Pu) dpm/g adjusted for the sample location on macroplot 1, as a function of soil particle diameter (D), cm, at various depths. The model used was ln Pu = $b_0 + b_1$ ln D

Depth (cm)	Intercept (b_0)	Slope (b_1)	Correlation coefficient (r)	Significant at $\alpha =$	n
0–3	5.62	−0.336	−0.312	0.01	72
3–6	4.40	−0.270	−0.291	0.05	69
6–9	1.69	−0.753	−0.471	0.001	50
9–12	2.40	−0.544	−0.564	0.001	69
12–15	1.47	−0.799	−0.719	0.001	52
15–18	0.583	−0.775	−0.706	0.001	47
18–21	0.375	−0.572	−0.358	—	22

DISCUSSION

A scenario of the contamination process based on these and other data is postulated The Pu-contaminated cutting oil, comparable to lightweight motor oil but often thinned by carbon tetrachloride, was stored in 55-gal barrels for up to 7 yr (1957–1964) Before placement in barrels, the oil was reportedly drained through 2–3-μm filters. Dilute hydrochloric acid formed by reaction of carbon tetrachloride and water may have led to the production of very low concentrations of plutonium chloride, a relatively soluble Pu compound (Cl76) Supporting this contention, a 0 01-μm filter removed only about 50% of the plutonium from similarly contaminated oil stored for shorter periods, indicating that much of the Pu was either monomeric or very small particles (Na76) However, during the long storage period, the Pu species remaining in the oil might have combined to form aggregates (Cl76) Unfortunately, the size and binding tenacity of these conglomerates, if formed, is unknown

Leakage from the barrels was most likely not large or fast at first, but may have become so with time. Contaminated oil was deposited onto the ground surface and stabilized the soil where plutonium became available for binding to soil particles Plutonium deposited as metal particles likely oxidized slowly at normal temperatures in the presence of air The resulting plutonium

oxide was relatively soluble in water compared to high-fired oxides but relatively insoluble in water compared to most metallic oxides. If plutonium chloride were deposited on the soil, it was likely hydrolyzed soon after first contact with water and eventually became oxidized. The solubility of these compounds was again probably low relative to most compounds but greater than the high-fired oxides (Cl76).

It is probable that all PuO_2 particles, or molecules, eventually became attached to soil particles. Most likely, this attachment took the form of easily erodible, agglomerated particles, each containing numerous PuO_2 and soil particles

Redistribution of contaminated soil from the various drum leakage events was probably a relatively short or erratic process occurring with surface disturbance and high winds, as indicated by the S-8 air sampler data The regressions of Pu concentration as a function of X or Y distance and the multiple regression including the same data indicated that the slope associated with the Y (south) term was steeper than the slope of the X (east) variable. Since the X term is primarily in the direction of the predominant wind and the Y direction is subjected mostly to downward slope, wind seems to be the more likely transport force.

The S-8 air data, coupled with prevailing wind information, and the regression of plutonium concentration vs distances east and south of the source are strong evidence that wind was the primary mode of plutonium dispersion from the oil barrel storage area to the study macroplots.

In time, dispersed plutonium-contaminated soil particles no longer were significantly redistributed by wind. Wind, precipitation, and gravity may have caused particles to migrate from exposed surfaces downward into the soil, where they were sheltered by larger particles, litter, or vegetation Soil–plutonium particles may have gradually broken down by natural weathering processes, allowing the constituents to disperse on a microscale (i e on the order of a few centimeters). This concept is supported by results of autoradiographic studies of soil from Macroplot 1 (Mc78) that indicated that most contaminated particles are very small or single particles. Furthermore, this process is compatible with the high degree of spatial variability observed

In summary, the major facets of the scenario include· (1) either before or shortly after leakage onto the ground surface, the Pu contaminant was in the form of an oxide; (2) the Pu oxide became attached to soil particles, (3) gusty winds combined with periods of surface disturbance heterogeneously redistributed the particles to the east and southeast of the barrel storage area; and (4) the soil–Pu particles were eventually broken down by weathering and were dispersed laterally and downward into the soil profile

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